

AMENDMENTS TO THE CLAIMS

1. (currently amended) A method for controlling microbes selected from the group consisting of protozoa, bacteria, fungi, viruses, and combinations thereof, said method comprising contacting the microbe with at least one functional microbiocidal transition metal silicate selected from the group consisting of: (a) cupric silicates having a silica to copper ratio in the range of 1:0.34 to 1:5.15; (b) zinc silicates having a silica to zinc ratio in the range of 1:2 to 1:12; (c) silver silicates having a silica to silver ratio in the range of 1:15 to 1:19.5; (d) manganese silicates having a silica to manganese ratio in the range of 1:1 to 1:1.9; and (e) zirconium silicates having a silica to zinc ratio in the range of 1:0.77 to 1:2.9;

wherein said transition metal silicates are prepared by the process comprising the steps of

- (i) adding a transition metal salt solution to a soluble alkali silicate solution under acidic conditions to form a mixture;
- (ii) forming a precipitate of a transition metal silicate, and
- (iii) washing and drying the precipitate thus formed to obtain the transition metal silicate.

2. – 54. (cancelled).

55. (previously presented) The method as claimed in claim 1, wherein the microbe is contacted with cupric silicate which is prepared under acidic conditions, has a silica to copper ratio of 1:5.15, and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.5; (c) 2.3; (d) 2.0 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.2 and 39.7 having peak heights of 2128, 1593 and 1470, respectively.

56. (previously presented) The method as claimed in claim 1, wherein the microbe is contacted with cupric silicate which is prepared under neutral conditions, has a silica to copper ratio of 1:1, and exhibits the following characteristics: characteristic g values of the peaks as

obtained by the electron spin resonance spectrometer being (a) 3.1; (b) 2.3; (c) 2.0; (d) 1.2 and (e) 0.9; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.7 having peak heights of 940, 764 and 694, respectively.

57. (previously presented) The method as claimed in claim 1, wherein the microbe is contacted with cupric silicate which is prepared under acidic conditions, has a silica to copper ratio of 1:0.78 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.2 and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16, 32 and 39 having peak heights of 835, 706 and 502, respectively.

58. (previously presented) The method as claimed in claim 1, wherein the microbe is contacted with cupric silicate which is prepared under extreme acidic conditions, has a silica to copper ratio of 1:0.53 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.1, (b) 2.0 and (c) 2.1; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.71 having peak heights of 400, 394 and 330, respectively.

59. (previously presented) The method as claimed in claim 1, wherein the microbe is contacted with cupric silicate which is prepared under extreme acidic conditions, has a silica to copper ratio of 1:0.34 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.1, and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.3 and 39.8 having peak heights of 541, 414 and 365, respectively.

60. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with zinc silicate which is prepared under neutral conditions, has a silica to zinc ratio of 1:12.13 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 5.4; (b) 4.5; (c) 2.5; (d) 2.1 and (e) 2.0; and X-ray

diffraction analysis having 3 significant peaks at 32.7, 59.5 and 26.2 having peak heights of 444, 307 and 263, respectively.

61. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with zinc silicate which is prepared under extreme acidic conditions, has a silica to zinc ratio of 1:2.46 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.0; (c) 2.5; (d) 1.8 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 11.0, 33.5 and 32.8 having peak heights of 2079, 835 and 664, respectively.

62. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with silver silicate which is prepared under neutral conditions, has a silica to silver ratio of 1:19.57 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.3; (c) 3.9 and (d) 2.0; and X-ray diffraction analysis having 3 significant peaks at 32.2, 46.2 and 27.8 having peak heights of 3945, 2421 and 1835, respectively.

63. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with silver silicate which is prepared under extreme acidic conditions, has a silica to silver ratio of 1:1.04 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.0 and (c) 1.9; and X-ray diffraction analysis having 3 significant peaks at 29.3, 47.6 and 42.3 having peak heights of 2217, 684 and 674, respectively.

64. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with manganese silicate which is prepared under neutral condition, has a silica to manganese ratio of 1:1.94 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 1.93 and (b) 2.06; and X-ray diffraction analysis having 1 significant peak at 30.6 having peak height of 148.0.

65. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with manganese silicate which is prepared under extreme acidic conditions, has a silica to manganese ratio of 1:1.09 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.1; (c) 2.1; (d) 2.1; (e) 2.0 and (f) 1.9; and X-ray diffraction analysis having 1 significant peak at 24.6 having peak height of 32.8.

66. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with zirconium silicate which is prepared under neutral conditions, has a silica to zirconium ratio of 1:2.9 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.42; (b) 1.82; (c) 2.24; (d) 2.3; (e) 2.18 and (f) 1.23;

67. (withdrawn) The method as claimed in claim 1, wherein the microbe is contacted with zirconium silicate which is prepared under extreme acidic condition, has a silica to zirconium ratio of 1:0.77 and exhibits the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.8; (c) 1.9; (d) 1.2; (e) 1.0 and (f) 0.9; and X-ray diffraction analysis having 1 significant peak at 10.8 having peak height of 84.80.

68. (cancelled).

69. (currently amended) The method as claimed in claim 1 [[68]], wherein the bacteria is selected from the group consisting of coliform bacteria, Gram positive bacteria, Gram negative bacteria, or a combination thereof.

70. (currently amended) The method as claimed in claim 1 [[68]], wherein the protozoa is *Cryptosporidium parvum*.

71. (currently amended) The method as claimed in claim 1 [[68]], wherein the fungus is a pathogenic fungus selected from the group consisting of *Sclerotium rolfsii*, *Rhizoctonia solani*, *Fusarium oxysporum*, *Pyricularia oryzae*, *Aspergillus* sps, or a combination thereof.

72. (cancelled).

73. – 77. (cancelled).

78. (currently amended) A method for controlling microbes selected from the group consisting of protozoa, bacteria, fungi, viruses, and combinations thereof, said method comprising contacting the microbe with at least one functional microbiocidal transition metal silicate selected from the group consisting of: (a) cupric silicates having a silica to copper ratio of 1:0.34, of 1:0.53, of 1:0.78, of 1:1, or of 1:5.15; (b) zinc silicates having a silica to zinc ratio in the range of 1:2 to 1:12; (c) silver silicates having a silica to silver ratio in the range of 1:15 to 1:19.5; (d) manganese silicates having a silica to manganese ratio in the range of 1:1 to 1:1.9; and (e) zirconium silicates having a silica to zinc ratio in the range of 1:0.77 to 1:2.9;

wherein said transition metal silicates are prepared by the process comprising the steps of

- (i) adding a transition metal salt solution to a soluble alkali silicate solution under acidic conditions to form a mixture;
- (ii) forming a precipitate of a transition metal silicate, and
- (iii) washing and drying the precipitate thus formed to obtain the transition metal silicate.

79. (new) A method for controlling microbes selected from the group consisting of protozoa, bacteria, fungi, viruses, and combinations thereof, said method comprising contacting the microbe with at least one microbiocidal transition metal silicate selected from the group consisting of: (a) cupric silicates having a silica to copper ratio in the range of 1:0.34 to 1:5.15; (b) zinc silicates having a silica to zinc ratio in the range of 1:2 to 1:12; (c) silver silicates having

a silica to silver ratio in the range of 1:15 to 1:19.5; (d) manganese silicates having a silica to manganese ratio in the range of 1:1 to 1:1.9; and (e) zirconium silicates having a silica to zinc ratio in the range of 1:0.77 to 1:2.9.

80. (new) The method for controlling microbes selected from the group consisting of protozoa, bacteria, fungi, viruses, and combinations thereof according to claim 79, wherein said method comprises contacting the microbe with at least one microbiocidal transition metal silicate selected from the group consisting of:

- i) cupric silicate having a silica to copper ratio of 1:5.15, and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.5; (c) 2.3; (d) 2.0 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.2 and 39.7 having peak heights of 2128, 1593 and 1470, respectively;
- ii) cupric silicate having a silica to copper ratio of 1:1, and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 3.1; (b) 2.3; (c) 2.0; (d) 1.2 and (e) 0.9; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.7 having peak heights of 940, 764 and 694, respectively;
- iii) cupric silicate having a silica to copper ratio of 1:0.78 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.2 and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16, 32 and 39 having peak heights of 835, 706 and 502, respectively;
- iv) cupric silicate having a silica to copper ratio of 1:0.53 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.1, (b) 2.0 and (c) 2.1; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.71 having peak heights of 400, 394 and 330, respectively;
- v) cupric silicate having a silica to copper ratio of 1:0.34 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance

spectrometer being (a) 2.1, and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.3 and 39.8 having peak heights of 541, 414 and 365, respectively;

vi) zinc silicate having a silica to zinc ratio of 1:12.13 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 5.4; (b) 4.5; (c) 2.5; (d) 2.1 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 32.7, 59.5 and 26.2 having peak heights of 444, 307 and 263, respectively;

vii) zinc silicate having a silica to zinc ratio of 1:2.46 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.0; (c) 2.5; (d) 1.8 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 11.0, 33.5 and 32.8 having peak heights of 2079, 835 and 664, respectively;

viii) silver silicate having a silica to silver ratio of 1:19.57 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.3; (c) 3.9 and (d) 2.0; and X-ray diffraction analysis having 3 significant peaks at 32.2, 46.2 and 27.8 having peak heights of 3945, 2421 and 1835, respectively;

ix) silver silicate having a silica to silver ratio of 1:1.04 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.0 and (c) 1.9; and X-ray diffraction analysis having 3 significant peaks at 29.3, 47.6 and 42.3 having peak heights of 2217, 684 and 674, respectively;

x) manganese silicate having a silica to manganese ratio of 1:1.94 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 1.93 and (b) 2.06; and X-ray diffraction analysis having 1 significant peak at 30.6 having peak height of 148.0;

xi) manganese silicate having a silica to manganese ratio of 1:1.09 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.1; (c) 2.1; (d) 2.1; (e) 2.0 and (f) 1.9; and X-ray diffraction analysis having 1 significant peak at 24.6 having peak height of 32.8;

xii) zirconium silicate having a silica to zirconium ratio of 1:2.9 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.42; (b) 1.82; (c) 2.24; (d) 2.3; (e) 2.18 and (f) 1.23; and

xiii) zirconium silicate having a silica to zirconium ratio of 1:0.77 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.8; (c) 1.9; (d) 1.2; (e) 1.0 and (f) 0.9; and X-ray diffraction analysis having 1 significant peak at 10.8 having peak height of 84.80.

81. (new) A transition metal silicate selected from the group consisting of:

- (a) cupric silicates having a silica to copper ratio in the range of 1:0.34 to 1:5.15;
- (b) zinc silicates having a silica to zinc ratio in the range of 1:2 to 1:12;
- (c) silver silicates having a silica to silver ratio in the range of 1:15 to 1:19.5;
- (d) manganese silicates having a silica to manganese ratio in the range of 1:1 to 1:1.9;

and

- (e) zirconium silicates having a silica to zinc ratio in the range of 1:0.77 to 1:2.9.

82. (new) A transition metal silicate according to claim 81, selected from the group consisting of:

i) cupric silicate having a silica to copper ratio of 1:5.15, and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.5; (c) 2.3; (d) 2.0 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.2 and 39.7 having peak heights of 2128, 1593 and 1470, respectively;

ii) cupric silicate having a silica to copper ratio of 1:1, and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 3.1; (b) 2.3; (c) 2.0; (d) 1.2 and (e) 0.9; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.7 having peak heights of 940, 764 and 694, respectively;

iii) cupric silicate having a silica to copper ratio of 1:0.78 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.2 and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16, 32 and 39 having peak heights of 835, 706 and 502, respectively;

iv) cupric silicate having a silica to copper ratio of 1:0.53 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.1, (b) 2.0 and (c) 2.1; and X-ray diffraction analysis having 3 significant peaks at 16.1, 32.2 and 39.71 having peak heights of 400, 394 and 330, respectively;

v) cupric silicate having a silica to copper ratio of 1:0.34 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 2.1, and (b) 2.0; and X-ray diffraction analysis having 3 significant peaks at 16.2, 32.3 and 39.8 having peak heights of 541, 414 and 365, respectively;

vi) zinc silicate having a silica to zinc ratio of 1:12.13 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 5.4; (b) 4.5; (c) 2.5; (d) 2.1 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 32.7, 59.5 and 26.2 having peak heights of 444, 307 and 263, respectively;

vii) zinc silicate having a silica to zinc ratio of 1:2.46 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.0; (c) 2.5; (d) 1.8 and (e) 2.0; and X-ray diffraction analysis having 3 significant peaks at 11.0, 33.5 and 32.8 having peak heights of 2079, 835 and 664, respectively;

viii) silver silicate having a silica to silver ratio of 1:19.57 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.3; (c) 3.9 and (d) 2.0; and X-ray diffraction analysis having 3 significant peaks at 32.2, 46.2 and 27.8 having peak heights of 3945, 2421 and 1835, respectively;

ix) silver silicate having a silica to silver ratio of 1:1.04 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance

spectrometer being (a) 4.3; (b) 4.0 and (c) 1.9; and X-ray diffraction analysis having 3 significant peaks at 29.3, 47.6 and 42.3 having peak heights of 2217, 684 and 674, respectively;

x) manganese silicate having a silica to manganese ratio of 1:1.94 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 1.93 and (b) 2.06; and X-ray diffraction analysis having 1 significant peak at 30.6 having peak height of 148.0;

xi) manganese silicate having a silica to manganese ratio of 1:1.09 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 4.1; (c) 2.1; (d) 2.1; (e) 2.0 and (f) 1.9; and X-ray diffraction analysis having 1 significant peak at 24.6 having peak height of 32.8;

xii) zirconium silicate having a silica to zirconium ratio of 1:2.9 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.42; (b) 1.82; (c) 2.24; (d) 2.3; (e) 2.18 and (f) 1.23; and

xiii) zirconium silicate having a silica to zirconium ratio of 1:0.77 and exhibiting the following characteristics: characteristic g values of the peaks as obtained by the electron spin resonance spectrometer being (a) 4.3; (b) 2.8; (c) 1.9; (d) 1.2; (e) 1.0 and (f) 0.9; and X-ray diffraction analysis having 1 significant peak at 10.8 having peak height of 84.80.